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P. Davis, T. Doeppner, J. R. Rygg, C. Fortmann, W.  
Unites, J. Salmonson, G. W. Collins, O. L. Landen, R.  
W. Falcone, S. H. Glenzer

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## MEASURE-RAY THOMSON SCATTERING EXPERIMENTS FROM SHOCK-COMPRESSED DEUTERIUM

P. Davis<sup>a,b</sup>, T. Doeppner<sup>b</sup>, J.R. Rygg<sup>b</sup>, C. Fortmann<sup>b</sup>, W. Unites<sup>b</sup>,  
J. Salmonson<sup>b</sup>, G.W. Collins<sup>b</sup>, O.L. Landen<sup>b</sup>, R.W. Falcone<sup>a</sup>  
and S.H. Glenzer<sup>b</sup>

<sup>a</sup>*University of California, Berkeley. Berkeley, CA, 94720, USA*

<sup>b</sup>*Lawrence Livermore National Laboratory. Livermore, CA 94551, USA*

**Abstract.** X-ray Thomson scattering has recently been shown to be an effective method of diagnosing a variety of high energy density plasma conditions. We apply this powerful technique to the widely studied problem of shock-compressed liquid deuterium. The behavior of deuterium under extreme conditions has received considerable attention due to its central role in models of giant planets and the importance of the high-pressure insulator-metal transition. We have used spectrally resolved x-ray scattering from electron-plasma waves to perform microscopic observations of ionization during compression. In these experiments, a single shock was launched in cryogenic deuterium reaching compressions of 3x. The 2 keV Ly- $\alpha$  line in silicon was used as an x-ray source in a forward scattering geometry. In addition to elastic scattering from tightly bound electrons, this low probe energy accessed the collective plasmon oscillations of delocalized electrons. Inelastic scattering from the plasmons allowed accurate measurements of the free electron density through the spectral position of the resonance and provided an estimate of the temperature through its ratio with the elastic feature. Combined with velocity interferometry from the reflective shock front, this lead to a direct determination of the ionization state. We compare the measured ionization conditions with computational models. Additionally, we discuss the possibility of using this technique to determine electrical conductivity and to directly observe pressure-induced molecular dissociation along the Hugoniot.

**Keywords:** Plasma Diagnostic, Dense Plasma Thomson Scattering, Deuterium Equation of State

**PACS:** 52.25 Jm, 52.35 Fp, 52.50 Lp

## INTRODUCTION

An understanding of the high-pressure behavior of hydrogen has been of fundamental interest since the 1935 prediction of high-pressure metallization by Wigner and Huntington<sup>1</sup>. In recent years, dynamic laser-driven experiments have led to the observation of the finite-temperature insulator-metal transition near 100 GPa.<sup>2</sup>

Continuing experimental work on a variety of platforms<sup>3-7</sup> and increasingly sophisticated *ab initio* calculations<sup>8-11</sup> have refined understanding of the high-pressure deuterium equation of state. However, strong interest in the behavior of warm, dense hydrogenic systems remains, thanks to applications to inertial confinement fusion<sup>12</sup> and planetary structure<sup>13</sup> as well as postulated high-pressure phase transitions.<sup>14,15</sup>

Experimental progress has been driven by the realization of extreme conditions in dynamic experiments. The high pressures reached in strong shocks are typically probed by diagnostics that make use of velocimetry measurements; determining shock and particle velocities allows closure of the Hugoniot relations and hence constrains the equation of state.<sup>5</sup> In addition, optical reflectivity has been used to infer the onset of metallization.<sup>16</sup>

Spectrally resolved x-ray scattering is a diagnostic that allows the microscopic properties of dense plasmas to be measured directly.<sup>17</sup> Here, we present measurements of the emergence of plasmon oscillations in shock-compressed deuterium, allowing an accurate determination of the free electron population, estimates of electron temperature and when coupled with VISAR, average ionization state  $Z$ .


## X-RAY THOMSON SCATTERING

X-ray Thomson scattering has emerged in recent years as a new way of probing a wide set of properties of dense plasmas. For example, recent experiments have used spectrally resolved forward scattering to detect pressure induced phase transitions in lithium hydride<sup>18</sup> and many-body effects in compressed boron.<sup>19</sup> The power of x-ray scattering as a diagnostic lies in the dependence of the scattering cross section on electronic and ionic structural quantities. For the Thomson cross-section  $\sigma_{\text{Th}}$  and incident and scattered wave-vectors  $k_0$  and  $k_s$ , the double differential cross section of x-ray scattering can be expressed as



(1)


The structure factor of dense plasmas can in turn be written<sup>20</sup>



(2)

using the model of a liquid metal. In this expression, the first term corresponds to scattering from electrons closely following ionic motion; the second corresponds to scattering from free electrons and the third to transitions of electrons from bound states into the continuum.

The nature of the inelastic scattering spectrum is described by the scattering parameter  $\alpha=1/k\lambda_s$ , where  $k$  is the scattering vector defined by the experimental geometry and in the strongly degenerate case of partially ionized deuterium, the screening length may be approximated by the Thomas-Fermi length,



(3)

In order to detect plasmon oscillations, we require probe scale lengths that extend beyond the screening length of the plasma, and hence  $\alpha>1$ .

Given the relatively low electron density of partially ionized deuterium and the need to detect small plasmon shifts associated with the early onset of ionization, we have developed a relatively soft, high-efficiency x-ray source line. The Lyman- $\alpha$  doublet of Silicon at 2005 eV shown in Figure 1 (a) provides a narrow-band, high contrast x-ray probe ideal for resolving plasmon features in deuterium at low ionization.<sup>21</sup> We have shown conversion efficiency from laser to x-ray emission on the order of  $10^{-3}$ , sufficient to provide  $10^{12}$  photons on target from a 200 J laser pulse, and good signal-to-noise in the inelastic scattering spectra.

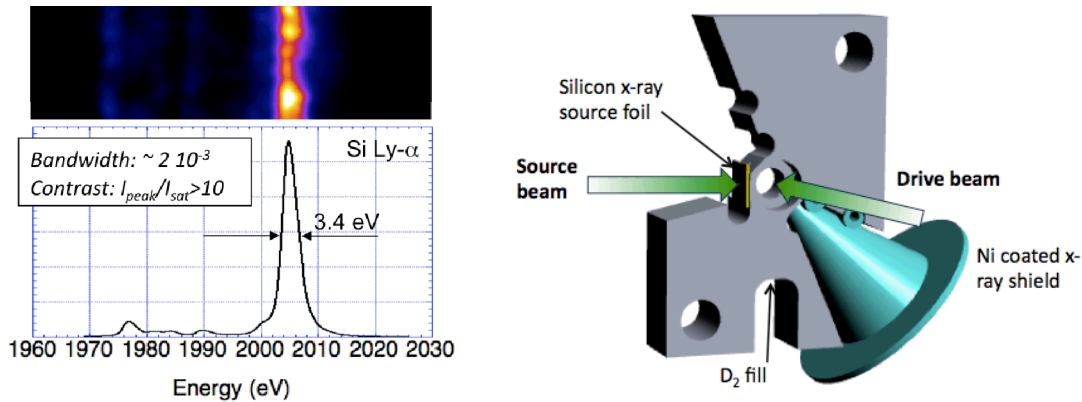
## EXPERIMENTS

Experiments were performed on the Janus laser at Lawrence Livermore National Laboratory, a two beam Nd:Glass facility. Samples were shocked with a 2 ns, frequency-doubled pulse. Using a 600  $\mu\text{m}$  phase plate, intensities of 40 TW/cm<sup>2</sup> were reached on target. A similar beam was used to pump the Si Ly- $\alpha$  resonance, propagating x-rays into the target perpendicular to the shock front.

Liquid deuterium was held at 19 K in a copper cell, shown schematically in Figure 1 (b). The front of the target was sealed with a layered 10  $\mu\text{m}$  CH ablator / 40  $\mu\text{m}$  Al pusher that minimized target preheat and drive laser bleed-through. The rear seal was an angled 130  $\mu\text{m}$  quartz window, allowing an optical view for the VISAR probe beam. 400  $\mu\text{m}$  x-ray access ports were bored perpendicular to the axis of the shock. One was covered with a Si<sub>3</sub>N<sub>4</sub> x-ray source foil, where probe light entered the target. The collection port was shielded with a Ni-coated plastic cone, eliminating contamination from x-ray emission driven by both lasers. The forward scattering port was situated at a  $\theta=45^\circ$  angle to the incoming x-rays, producing a scattering vector



(4)



**FIGURE 1.** (a) Silicon Lyman- $\alpha$  x-ray probe spectrum. This narrow band, high efficiency source allows plasmons to be observed at even small fractional ionizations in D<sub>2</sub>. (b) Schematic of target cell

for cryogenic deuterium. The cell defines a scattering angle of  $45^\circ$ . X-ray signal is shielded from background by Ni cone.

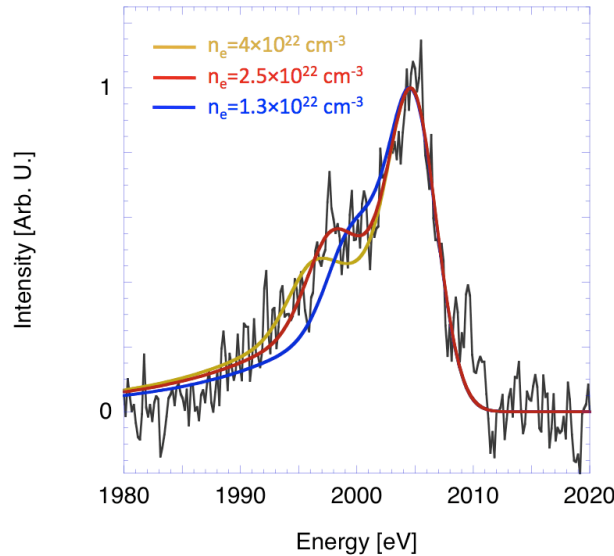
For an average ionization of  $Z=0.1$ , a  $3\times$  compressed deuterium plasma has an electron density of  $n_e=1.5 \times 10^{22} \text{ cm}^{-3}$ , with a scattering parameter of  $\alpha=1.3$  at  $\theta=45^\circ$ . The forward scattering diagnostic is thus strongly collective, allowing the detection of plasmons at even very weakly ionized conditions.

The shock conditions of the target were monitored with a VISAR diagnostic. Deuterium shock velocities can be extracted directly from the motion of the reflecting shock, made possible by the increase in reflectivity due to the onset of ionization in the shock front.<sup>22</sup> This procedure allows a precise synchronization of the x-ray probe to the shock in time and space. Moreover, it allows the plasma pressure and mass density to be inferred using the Hugoniot relations and the equation of state.

A forward scattered x-ray spectrum is shown in Figure 2. Here the feature at  $E_0=2005 \text{ eV}$  corresponds to elastic scattering from bound electrons or those closely following nuclear motion. The inelastic feature  $7 \text{ eV}$  downshifted from  $E_0$  is due to free-free plasmon oscillations. The spectral position of this feature is closely linked to the free electron density as can be seen in the approximate plasmon dispersion relation

$$\hbar\omega = \hbar\omega_p + \frac{1}{2}m_e v_{th}^2 \left( \frac{2\pi}{\lambda} \right)^2 \quad (5)$$

where  $\omega_p$  is the plasma frequency,  $v_{th}$  is the thermal velocity and  $\lambda$  is the thermal wavelength.

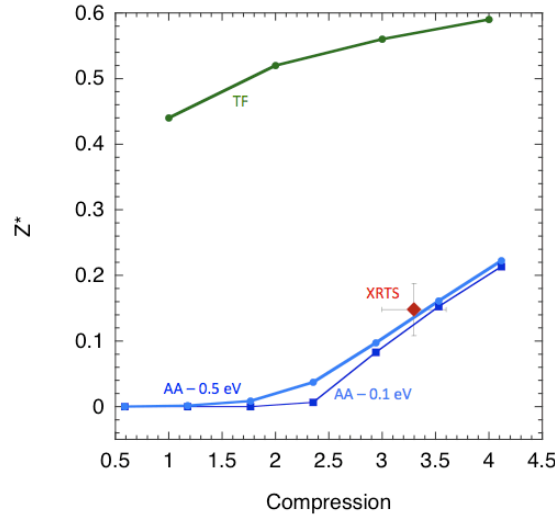


**FIGURE 2.** Measured forward scattering spectrum with theoretical fits corresponding to densities between  $1.3$  and  $4 \times 10^{22} \text{ cm}^{-3}$ . Combined with a determination of mass density from VISAR, this allows a direct measure of average ionization state.

We show three theoretical fits that incorporate collisional effects in the Born-Mermin approximation for  $S_{ee}^0(k, \omega)$ .<sup>23</sup> These fits correspond to densities of 1.3, 2.5 and  $4 \times 10^{22} \text{ cm}^{-3}$ . The best fit of the plasmon is  $2.5 \times 10^{22} \text{ cm}^{-3}$  with 15% uncertainty due mostly to noise in the spectrum. All three curves correspond to a temperature of 0.15 eV using a screened one-component plasma model for  $S_{ii}(k, \omega)$ .<sup>24</sup> Shock velocity at the time of the x-ray pulse was measured to be 12.3 km/s. From this we infer a pressure of 18.5 GPa and compression of  $3.3\times$  using Hugoniot data from recent *ab initio* calculations.<sup>11</sup> Combining the measured electron and mass densities, we extract the average ionization as  $Z=0.15\pm0.04$ .

## RESULTS

The transition from insulating liquid to metal is driven by pressure-induced ionization and is quite abrupt. In Figure 3 we plot our measurement with theoretical curves for ionization as a function of compression using average-atom and Thomas-Fermi approximations, common models for equation of state and hydrodynamic calculations. AA calculations are performed at  $T_e=0.1$  and 0.5 eV, showing minimal variations with temperature. We also note that measured ionizations are consistent with the reflectivities previously observed by Celliers and coworkers.<sup>16</sup>



**FIGURE 3.** Ionization state calculated as a function of compression using average-atom (AA) and Thomas Fermi (TF) models shown with the measured value using Thomson scattering and VISAR.

State-of-the-art calculations in the moderate compression regime rely on quantum molecular dynamics models, which use physical picture frameworks that avoid defining precise ionization states. Under truly warm, dense conditions like this semi-conducting deuterium liquid, this makes comparison to electron density measurements challenging.

However, it is possible to extract optical properties from QMD calculations using the Kubo-Greenwood formalism for calculating dynamic conductivities.<sup>25</sup> Since the electron-electron structure factor is related to the complex dielectric function through the fluctuation-dissipation theorem, ongoing research is aimed at using

scattering results to validate such calculations at the level of the dielectric function, rather than bulk averaged quantities.

## CONCLUSION

Collective x-ray Thomson scattering has been observed in laser-driven deuterium for the first time. In particular, we have detected the emergence of free electron oscillations due to pressure ionization. Coupled with velocimetry, this allows an accurate determination of the ionization fraction, a quantity that reflects the microscopic physics behind the onset of metallization. Future work will extend this method to the determination of quantities of fundamental interest such as the dynamic conductivity.

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